

Standard Form 298 (Rev. 2-89)
Prescribed by ANSI Std. Z39-18

AFOSR DURIP-96 Award #F49620-96-1-0372

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Development of a Spatiotemporal Spin Resonance NSOM for Studies of Magnetic Nanostructures

FINAL TECHNICAL REPORT

I) Equipment

We have successfully constructed a top-loading variable-temperature (1K - 300K) magneto-optical (0-7T) near-field scanning optical microscope which is capable of examining thin films and quantum structures with a spatial resolution of ~ 80 nm and a temporal resolution of 100 femtoseconds. Spatio-temporal measurement methods include photoluminescence (PL) and absorption spectroscopy/imaging, where high resolution (~ 70 μ eV) spectra are obtained rapidly at any point on the surface (or below the surface) using nitrogen-cooled CCD arrays, and appropriate spatial images collected at a desired detection energy. This has been made possible by purchasing a femtosecond tunable laser system capable of generating an ultrafast white-light continuum within this time scale. The costs for this project are divided as follows:

(a) AFOSR DURIP Award: \$90,000 Coherent Laser Model 9000 Regenerative Amplifier and Coherent Laser Model 9400 Optical Parametric Amplifier System. Equipment to enable full optical imaging spectroscopies within femtosecond time scales using an ultrafast white-light source.

(b) UCSB Cost Sharing: \$30,000 Design, Construction, and Operation of Titanium Low-temperature Near-field Scanning Optical Microscope for top loading magneto-optical cryostat, including computers, optics, and electronics.

II) Summary of Research Projects

- *II-VI quantum dots, magnetic quantum dots, and strain-induced quantum dots*

We have recently succeeded in the fabrication and *direct* observation of 0D exciton confinement in *wide-gap* II-VI quantum dots. These nanostructures are formed during the strained layer epitaxy of (cubic) CdSe ($E_g = 1.75$ eV) on ZnSe ($E_g = 2.8$ eV) with a lattice mismatch $\sim 7\%$. The 0D nature of the confined electronic states in these quantum dots is directly revealed through PL spectroscopy with high spatial resolution (~ 100 nm) carried out using the low-temperature near-field scanning optical microscope. The smooth, inhomogeneously broadened lineshape of typical far field PL spectra is seen to evolve into a spectrum characterized by sharp, resolution-limited (0.8 meV) reproducible spectral features in the near-field, arising from a convolution of the delta-function density-of-states of each individual quantum dot region and an envelope determined by the statistical distribution of quantum dots. Measurements in II-VI CdSe quantum dots electronically coupled to adjacent MnSe magnetic layers have revealed the existence of magnetic quantum dots, exhibiting field-tunable Zeeman splittings in individual dots. These near-field optical spectra at low temperatures demonstrate the ability to fabricate and measure genuine "spin quantum dots" in applied magnetic fields, and explore spin-dependent phenomena in zero-dimensional structures. The observation of 0D states in II-VI nanostructures opens up exciting possibilities for studying static and dynamic *spin dependent phenomena* in "quantum spin dots" by the incorporation of magnetic ions into II-VI nanostructures. Finally, another scheme entails the use of the strain field from overgrown CdSe quantum dots to produce 0D confinement regions in a magnetic quantum well below that is separated from the dots

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by a ZnSe spacer. Such strain-patterned dots have recently been very successfully observed in III-V nanostructures and are found to exhibit high quantization effects along with a low degree of inhomogeneous broadening. We have used the NSOM to image zero-dimensional (0D) excitonic confinement in locally strained $\text{Zn}_{1-x}\text{Cd}_x\text{Se}$ quantum wells. Strain fields from self-organized CdSe quantum dots locally modulate the band structure of a nearby quantum well (QW) in a heterostructure, resulting in confinement in all three dimensions. The 0D nature of excitonic confinement is verified by the observation of sharp lines in photoluminescence (PL) spectra. The temperature dependence of the PL lifetime is markedly different than that of the CdSe quantum dots, demonstrating tunneling from 0D to 2D electronic states. High-resolution spectra show that the PL lines from the localized states are split into linearly polarized doublets.

- *ferromagnetic patterning for magnetoelectronics*

In contrast to embedding magnetic moments within a semiconductor heterostructure, it is important to demonstrate an ability to guide spin-dependent transport using field gradients from magnetic films grown upon a semiconductor heterostructure. The spin-dependent properties of these magnetically-patterned nanostructures are resolved using both near-field scanning optical microscopy (NSOM) and magnetotransport studies. We have developed magnetically active II-VI diluted magnetic semiconductor (DMS) surface quantum well "substrates" upon which to epitaxially grow and pattern ferromagnetic Fe films in order to produce spin-dependent potentials. This material phase produces flux-focusing magnetic patterns which have included planar wedges and variable-spaced single domain particle arrays with nominally 0.1-0.5 micron feature sizes. These patterns are designed to create a field-driven spin-dependent energy landscape for optically-pumped or doped carriers and provide a basis for spatial measurements of electronic spin transport. We have grown a hybrid ferromagnetic-DMS quantum well structure by depositing a 50 nm thick epitaxial film of Fe on top of a single 12 nm quantum well containing 3 monolayers of Mn ions. This wet-etched single-crystal Fe film was processed using a newly developed chemical technique, *without any electronic degradation of the underlying semiconductor heterostructure*. The magnetic field due to the iron penetrates the quantum well near the edges of the patterned regions. A combination of NSOM photoluminescence, far-field Hanle effect, and time-resolved Faraday rotation measurements have been performed in and around these structures which demonstrate that the films can be processed without significantly altering the electronic properties of the quantum well.

- *NSOM of single and multiple InGaN quantum wells*

We have used near-field scanning optical microscopy to study photoluminescence (PL) in single and multiple InGaN/GaN quantum wells. To our knowledge, these represent the first high-resolution spatial images of PL from a InGaN structures over a broad range of temperatures. Measurements have been made for temperatures between 50 and 295 K with spatial resolution of approximately 100 nm. One of the principal advantages of this technique is the capability of correlating spatially-resolved PL with topographic information, and the work is aimed at identifying the mechanism of strong optical activity in nominally very disordered material. All of the structures were grown at UCSB using MOCVD methods under the supervision of Professor Evelyn Hu. To our surprise, the strongest (~ 50 %) spatial modulation in the PL intensity occurs in the vicinity of large (~ 500 nm diameter) pits in the heterostructure. Weaker (~15 %) variations in the PL do not correlate directly with the presence of pits. In contrast, the green band luminescence (analogous to the yellow band in GaN) appears only when the sample is excited over the pits. A particularly important observation is that the spectrum of the quantum well PL shows no significant variations on the length scales probed in our experiment. We therefore find no evidence for the recombination of localized carriers (or quantum dots) in the InGaN/GaN system at temperatures above 50 K, despite several claims in the literature.

- *Publications using the new instrument*

1. F. S. Flack, A. Hunt, H. Hennessey, N. Samarth, J. Levy, V. Nikitin, S. A. Crooker, D. D. Awschalom, "Growth Dynamics and Exciton Localization in Strained CdSe Quantum Structures," in *Optoelectronic Materials: Ordering, Composition Modulation, and Self-Assembled Structures*, ed. by E. D. Jones, A. Mascarenhas and P. Petroff [Materials Research Society Symposium Proceeding, Pittsburgh, 1996], Vol. 417, p. 169.
2. N. Samarth and D. D. Awschalom, "Spin Coherence and Imaging in Magnetic Quantum Systems," Proceedings of the International Conference on Quantum Circuits and Devices, Alexandria, Egypt, June 1996, Eds. K. Ismail and S. Bandopadhyay [World Scientific, Singapore, 1997], p. 136.
3. V. Nikitin, P. A. Crowell, J. Levy, F. Flack, N. Samarth, and D. D. Awschalom, "Near-field Optical Spectroscopy of Magnetic Semiconductor Quantum Dots," Proceedings of the 23rd International Conference on the Physics of Semiconductors, Berlin, Germany, July 21-26, 1996 [World Scientific Press, Singapore, 1996], Vol. 2, p. 1441.
4. P. A. Crowell, V. Nikitin, J. Levy, F. Flack, N. Samarth, and D. D. Awschalom, "Spatiotemporal Near-field Spin Spectroscopy in Digital Magnetic Heterostructures," Proceedings of the 23rd International Conference on the Physics of Semiconductors, Berlin, Germany, July 21-26, 1996 [World Scientific Press, Singapore, 1996], Vol. 3, p. 2023.
5. P. A. Crowell, V. Nikitin, D. D. Awschalom, F. Flack, N. Samarth, A. C. Marley, S. Gider, S. S. P. Parkin, and G. A. Prinz, "Magneto-optical Spin Spectroscopy in Hybrid (Ferro)Magnetic Semiconductor Heterostructures," *J. Appl. Phys.* **81**, 5441 (1997).
6. F. Flack, V. Nikitin, P. Crowell, J. Shi, J. Levy, N. Samarth, and D. D. Awschalom, "Near-Field Optical Spectroscopy of Localized Excitons in Strained CdSe Quantum Dots," *Phys. Rev. B* **54**, Rapid Communications, R17312 (1996).
7. V. Nikitin, P. A. Crowell, J. A. Gupta, D. D. Awschalom, F. Flack, and N. Samarth, "Zero-Dimensional Excitonic Confinement in Locally Strained ZnCdSe Quantum Wells," *Appl. Phys. Lett.* **71**, 1213 (1997).
8. D. D. Awschalom and N. Samarth, "Spin Spectroscopy and Coherence in Magnetic Quantum Structures," in *Dynamical Properties of Unconventional Magnetic Systems*, NATO ASI Series Volume X, ed. A. T. Skjeltorp [Kluwer Academic, The Netherlands, 1997].
9. P. A. Crowell, V. Nikitin, J. A. Gupta, D. D. Awschalom, F. Flack, and N. Samarth, "Optical Spectroscopy of II-VI (Magnetic) Semiconductor Quantum Dots," *Physica E*, submitted (1997).
10. P.A. Crowell, D.K. Young, S. Keller, E.L. Hu, and D.D. Awschalom, "Near-Field Scanning Optical Spectroscopy of an InGaN Quantum Well", *Appl. Phys. Lett.*, submitted for publication (1997).